

### REMARKS

Applicants have amended their claims in order to further define various aspects of the present invention, and to simplify and facilitate the prosecution of the above-identified application. Specifically, the remaining pending claims being considered on the merits in the above-identified application have been reviewed and "for" clauses replaced. Note, for example, amendments to claims 1-3, 23, 28-32, 34 and 38. While Applicants disagree with the Examiner that the "for" clauses cause the claims to be indefinite, the present amendments have been made to facilitate prosecution of the above-identified application.

In addition, each of claims 1 and 23 has been amended to recite that the graphite powder manufactured is adapted to be an active material of a lithium negative electrode; and claims 30 and 38 have similarly been amended to recite that the graphite powder manufactured is adapted to be an active material of a lithium negative electrode. Claims 1 and 23 have each been further amended to recite that the pulverized graphite has a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure; claims 30 and 38 have correspondingly been amended to recite that the graphite powder provided, has a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure. Claims 1 and 23 have been further amended to recite that the graphite powder is heated as a heat treatment, and claim 23 has been

still further amended to recite the immersion, to transform the crystalline structure of the graphite powder to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure; claims 30 and 38 have correspondingly been amended to recite that the heating and immersing, or treating, of the graphite powder, is a treatment, such that the treated graphite powder has a fraction of a hexagonal structure of at least 80% by weight and a fraction of a rhombohedral structure of at most 20% by weight. In light of these amendments, and in order to facilitate proceedings, claims 4, 7-9, 33, 35-37 and 39 have been canceled without prejudice or disclaimer.

Applicants have added new claims 40-43 to the application. These new claims, dependent respectively on claims 1, 23, 30 and 38, recite that the graphite powder is in powder form when transformed to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure. These new claims 40-43 read on the elected invention and species.

The restriction requirement and election-of-species requirement, referred to in the first two paragraphs on page 2 of the Office Action mailed April 20, 2001, are noted. It is respectfully submitted that the presently amended independent claims, as well as claims dependent thereon, previously reading on the elected invention and elected species, still read on the elected invention and elected species.

The comment by the Examiner in lines 3 and 4 on page 2 of the Office Action mailed April 20, 2001, that the non-elected

subject matter (e.g., acid solution of claim 30, 23b, etc.) "should be removed from these claims" is noted. Applicants respectfully decline removing the non-elected non-elected species from the generic claims. To the contrary, it is respectfully submitted that the generic claims must be considered as a whole, after a conclusion of allowability of the subject matter of the elected species, and that both the elected and non-elected species should be allowed to issue in a U.S. patent based upon the present application.

Rejection of various of the claims presently being considered on the merits in the above-identified application, under the second paragraph of 35 USC 112, as set forth on page 2 of the Office Action mailed April 20, 2001, is noted. The claims, including claim 1, line 5, have been amended to delete "for" clauses; and, in particular, claim 1, line 5 has been amended to recite the step of sieving to recover graphite powder having a specified maximum particle diameter. In view thereof, it is respectfully submitted that the basis for rejection of the present claims, as set forth in Item A) on page 2 of the Office Action mailed April 20, 2001, is moot. It is respectfully submitted that claim 1 as presently amended clearly has antecedent basis for "the crystalline structure", in reciting that pulverized graphite having a specific crystalline structure is produced in the pulverizing step. In view of canceling of claim 4, it is respectfully submitted that the basis for rejection of claim 4, set forth in Item C) on page 2 of the Office Action mailed April 20, 2001, is moot.

Applicants respectfully submit that all of the claims presented for consideration by the Examiner patentably distinguish over the teachings of the references applied by the Examiner in rejecting the claims in the Office Action mailed April 20, 2001, that is, the teachings of the U.S. patents to Tsukagoshi, et al., No. 4,221,773, and to Bennett, et al., No. 5,672,327, and British Patent No. 1,087,046 (Longstaff), under the provisions of 35 USC 103.

It is respectfully submitted that these references as applied by the Examiner would have neither taught nor would have suggested such a method of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, as in the present claims, including the pulverizing of raw graphite to produce pulverized graphite having a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure; and heating the graphite powder as a heat treatment to transform the crystalline structure to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure. See claim 1; note also claim 23. In connection with claim 23, note also the immersing step, and it is respectfully submitted that the teachings of the applied references would have neither taught nor would have suggested such method as in claim 23, including the immersing step into an acidic solution and further processing to transform the crystalline structure of the graphite powder to at least 80%

by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure.

Moreover, it is respectfully submitted that the teachings of these applied references would have neither taught nor would have suggested such a method of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, as in the present claims, including providing graphite powder having a particle size equal to or smaller than 100  $\mu\text{m}$ , this graphite powder having a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure; and heating the graphite powder, or immersing the graphite powder, respectively as a heat treatment or into an acidic solution, to form treated graphite powder having a fraction of a hexagonal structure of at least 80% by weight and a fraction of a rhombohedral structure of at most 20% by weight. See claims 30 and 38.

Furthermore, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such a method of manufacturing graphite powder as in the present claims, having the recited heating to transform the crystalline structure to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure, and including the further heating step, at a higher temperature than the heat treatment to transform the crystalline

structure, to eliminate impurities (see claim 1, as well as the heating step of claim 23; note also claim 34).

In addition, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such a method of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, as in the present claims, wherein the temperature of the heat treatment to transform crystalline structure is at least 900°C (see claim 2); and wherein the temperature of the heat treatment to eliminate impurities is at least 2700°C (see claim 3). Note also claims 28 and 29, respectively further defining the temperature ranges for the heat treatment to transform crystalline structure and for the heat treatment to eliminate impurities.

Furthermore, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such a method of manufacturing graphite powder adapted to be an active material in a lithium battery negative electrode, as in the present claims, and wherein the graphite powder has a fraction of a hexagonal structure of at least 90% by weight (see claim 31) and has a fraction of a rhombohedral structure of at most 10% by weight (see claim 32); or wherein the graphite powder is in powder form in transforming the graphite to increased hexagonal structure (see claims 40-43).

The present invention is directed to a method of manufacturing graphite powder adapted to be used as negative

electrode active material in, e.g., a lithium battery, which manufactured carbon powder, when used in such negative electrode, has a high energy density and a long life, and which has excellent intercalation capacity (that is, an increased capacity of the battery).

Carbon material has been proposed as the negative electrode active material, in place of lithium metal, in lithium batteries, in which charge and discharge reactions involving lithium ion intercalation into the carbon material and deintercalation from the carbon material occur. However, in previously proposed lithium batteries using carbon material as active material for the negative electrode, a large capacity can not be obtained; moreover, preferable performances of rapid charging and discharging are not obtained. Note the paragraph bridging pages 2 and 3 of Applicants' Substitute Specification.

Against this background, Applicants provide a method for forming carbon powder to be used as active material for the negative electrode of a lithium battery, which active material formed achieves good charging and discharging, while having a large capacity (large intercalation capacity; that is, an increased capacity). Applicants have found that by limiting the amount of rhombohedral structure of the graphite powder, and increasing the amount of graphite powder having hexagonal structure, advantages are achieved. By reducing the graphite powder having rhombohedral structure and increasing the graphite powder having hexagonal structure, intercalation capacity of the negative electrode is increased. That is, the

number of storage sites for lithium atoms in the negative electrode is increased.

In accordance with the present invention, graphite is used as material for the negative electrode, and focus is on its crystallinity. That is, Applicants have found that by providing a specific ratio of hexagonal crystal to rhombohedral crystal, of the graphite powder of the negative electrode, improvements in capacity are achieved. It is respectfully submitted that in the present invention it is the crystal structure upon which focus is made in increasing intercalation capacity. Utilizing an amount of hexagonal crystal structure relative to rhombohedral crystal structure, unexpectedly better results of improved capacity are achieved, for the battery of the present invention.

Moreover, Applicants provide various techniques for achieving the amount of hexagonal crystalline structure, relative to rhombohedral crystalline structure, of the graphite powder according to the present invention, to achieve the advantages of the present invention. That is, Applicants have found that the reason for the small lithium intercalation-deintercalation capacity of conventional graphite powder is due to the fact that the crystalline structure of the graphite has been transformed to rhombohedral structure during processing in forming the graphite powder (e.g., during pulverizing) and has generated amorphous carbon, with the result that the lithium intercalation-deintercalation reaction is disturbed by the rhombohedral structure and the amorphous carbon. See the paragraph bridging pages 7 and 8 of



Applicants' Substitute Specification. Applicants provide techniques for decreasing the existing fraction of the rhombohedral structure and increasing the existing fraction of the hexagonal structure, in the graphite powder, so as to achieve graphite powder forming a negative electrode active material which can be used in a lithium battery to provide large load capacity and a high energy density. Note, for example, the first full paragraph on page 13 of Applicants' Substitute Specification. Note also the paragraph bridging pages 9 and 10, and the second and third paragraphs on page 10, of Applicants' Substitute Specification; and the second full paragraph on page 11 of Applicants' Substitute Specification.

In addition, Applicants have found that by removing impurities contained in the raw graphite powder, especially silicon, an improved negative electrode active material can be achieved, and provide a technique for removing the impurities, including heating at relatively high temperatures. Note the paragraph bridging pages 10 and 11 of Applicants' Substitute Specification.

Longstaff discloses graphite suitable for use in nuclear reactors, particularly well suited for fuel tubes in a high-temperature gas-cooled reactor, the graphite being in the form of an article wherein about 50% of the open porosity includes pores of size 5-10 microns, with substantially the remaining 50% of the open porosity being micropores of size less than 0.1 microns, there being few pores in the size range of 0.1-5 microns. Note page 1, lines 42-56. Also disclosed in this

British patent is a method for production of a graphite artefact, including taking a grist of nuclear grade graphite whereof substantially all the particles are of a size greater than about 50 microns, mixing and impregnating such powder with a binder to form a green mix, working such a mix to form a green artefact, baking such an artefact, impregnating such a baked artefact with a pitch impregnate and heating the baked, impregnated artefact to a graphitizing temperature. Note claim 3 of this patent; see also page 1, lines 64-76. Note also page 2, lines 27-33, describing use of the artefact for fuel tubes.

It is emphasized that Longstaff discloses graphite artefacts suitable for use in nuclear reactors. It is respectfully submitted that the method utilized for forming such graphite artefacts, in Longstaff, does not disclose, nor would have suggested, a method of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, as in the present claims; and, moreover, would have neither taught nor would have suggested the heating to transform the crystalline structure to at least 80% hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure, as in the present claims. Furthermore, it is respectfully submitted that the British patent document does not disclose pulverizing to produce pulverized graphite, or utilizing graphite powder, having the recited crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure; nor would have disclosed or

suggested desirability of providing powder having crystalline structure of at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure achieving an unexpectedly better negative electrode active material of a lithium battery, or the processing techniques (heating, immersing in acid) for achieving the crystalline structure having the specified amount of hexagonal crystalline structure and rhombohedral crystalline structure.

In addition, it is again emphasized that Longstaff is concerned with graphite structures suitable for use in nuclear reactors, in particular, for fuel tubes in a high-temperature gas-cooled reactor. Such disclosure would have neither taught nor would have suggested the desired treatment to provide the crystalline structure as in the present claims; or, moreover, the additional heating to eliminate impurities.

Tsukagoshi, et al. discloses a method of producing a diaphragm of an acoustic instrument, having a low density and a high elasticity, the method having the steps of blending and kneading carbon powders and a plastic, shaping the blend into a desired form, and carbonizing the shaped blend. See column 1, lines 63-68. Note also column 2, lines 24-29.

It is emphasized that Tsukagoshi, et al. is concerned with producing a diaphragm of an acoustic instrument; and it is respectfully submitted that this reference would have neither taught nor would have suggested a method for manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode. Moreover, it is respectfully submitted that Tsukagoshi, et al. does not

disclose, nor would have suggested, use of, e.g., pulverized raw graphite having a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure; unexpectedly better results as an active material of a lithium battery negative electrode for graphite powders having at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure; and the treatment techniques (heating or immersing) achieving such amounts of hexagonal crystalline structure and rhombohedral crystalline structure of the manufactured graphite powder according to the present invention.

It is respectfully submitted that the secondary reference applied by the Examiner in combination with Tsukagoshi, et al., that is, Bennett, et al., would not have rectified the deficiencies of Tsukagoshi, et al. such that the presently claimed subject matter as a whole would have been obvious to one of ordinary skill in the art. Bennett, et al. discloses a method of beneficiating kish graphite to high purity levels in an economic manner without undue destruction of the coarse graphite flakes. The kish graphite is described as a term for graphite in waste that forms during the iron production process and during initial steel making, containing graphite in a flake form, iron, lime-rich slag and other materials. Note particularly column 1, lines 41-46. As a method of recovering high purity flake graphite from kish, note column 2, lines 9-17. Note also column 3, lines 25-37.

Initially, note that Tsukagoshi, et al. is concerned with providing a carbon diaphragm; while Bennett, et al. is directed to providing coarse graphite flakes finding use in applications such as refractories, as a coating material in foundries, in brake linings, batteries, carbon brushes, pencils, propellants, as a precursor for expandible graphite and as a lubricant, as well as in other areas. It is respectfully submitted that one of ordinary skill in the art concerned with in Tsukagoshi, et al. would not have looked to the teachings of Bennett, et al. That is, in view of the different technologies involved in Tsukagoshi, et al. and in Bennett, et al., and different problems addressed by each, these two patents are in non-analogous arts.

Furthermore, it is respectfully submitted that there would have been no motivation for combining the teachings of Tsukagoshi, et al. and Bennett, et al., as applied by the Examiner, particularly in light of the different technologies involved in each, different materials formed and different problems addressed.

In any event, even assuming, arguendo, that the teachings of Tsukagoshi, et al. and Bennett, et al. were properly combinable, such combined teachings would have neither disclosed nor would have suggested, the presently claimed subject matter, including the manufacture of graphite powder adapted to be an active material of a lithium battery negative electrode; and/or wherein the pulverized graphite (graphite powder utilized at one stage of the method) has a crystalline structure which is at least 30% by weight rhombohedral

crystalline structure and at most 70% by weight hexagonal crystalline structure; desirability of utilizing graphite powder having at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure as the graphite powder to be an active material of the aforementioned negative electrode; and techniques for providing the graphite powder having the specified hexagonal and rhombohedral crystalline structures, as in the present claims.

According to the present invention, the pulverized powder is, for example, treated by a heat treatment at a stage of the process near completion of the process, to convert rhombohedral crystalline structure, e.g., generated by the pulverizing, to hexagonal crystalline structure. Note that each of Longstaff and Tsukagoshi, et al. provide articles (respectively a fuel tube and diaphragm of an acoustic instrument), not powder. Assuming, arguendo, that the articles of Longstaff and Tsukagoshi, et al. were pulverized to obtain powder, such pulverizing would destroy crystalline structure, and increase rhombohedral crystalline structure and decrease hexagonal crystalline structure, as seen by the present application. Clearly, the teachings of the applied references provide no suggestion in connection with the presently claimed method.

The contention by the Examiner that the claims do not exclude the additional steps and materials of the references, nor do the "for" clauses actually limit the claims, is noted. It must be emphasized, however, that Longstaff provides an

article, not graphite powder, which graphite powder must be given weight in connection with consideration of the present claims. Note especially claims 40-43. Moreover, the Examiner has not established that the teachings of the applied references would provide graphite powder having the recited crystalline structure, which must be given weight in determining patentability of the present claims.

In view of the foregoing comments and amendments, reconsideration and allowance of all claims remaining in the application are respectfully requested.

Attached hereto is a marked-up version of the changes made in the claims by the current Amendment. This marked-up version is on the attached pages, the first page of which is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE".

To the extent necessary, Applicants petition for an extension of time under 37 CFR § 1.136. Please charge any shortage in fees due in connection with the filing of this paper, including extension of time fees, to the Deposit Account No. 01-2135 (Case No. 503.34465VC3) and please credit any excess fees to such Deposit Account.

Respectfully submitted,

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS

Please cancel claims 4, 7-9, 33, 35-37 and 39 without prejudice or disclaimer, and amend the claims remaining in the application as follows:

1. (Amended) A method [for] of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, comprising the steps of:

pulverizing raw graphite, to produce pulverized graphite having a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure;

sieving said pulverized graphite [for obtaining] to recover graphite powder having a maximum particle diameter of 100  $\mu\text{m}$ ;

heating said graphite powder as a heat treatment [for transforming] to transform the crystalline structure to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure; and

further heating said graphite powder, at a higher temperature than said heat treatment [for transforming] to transform the crystalline structure, [for eliminating] to eliminate impurities.

2. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 1, wherein



the temperature of said heat treatment [for transforming] to transform crystalline structure to hexagonal structure is at least 900°C.

3. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 1, wherein

the temperature of said heat treatment [for eliminating] to eliminate impurities is at least 2700°C.

23. (Amended) A method [for] of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, comprising the steps of:

pulverizing raw graphite, to produce pulverized graphite having a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure;

sieving said pulverized graphite [for obtaining] to obtain graphite powder having a maximum particle diameter of 100  $\mu\text{m}$ ; and either

(a) heating said graphite powder as a heat treatment [for transforming] to transform the crystalline structure of said graphite powder to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure, and further heating said graphite powder, at a higher temperature than said heat treatment [for transforming] to transform the crystalline structure, [for eliminating] to eliminate impurities; or

(b) immersing said graphite powder into an acidic solution as an immersing treatment, washing with water, neutralizing and drying, to transform the crystalline structure of said graphite powder to at least 80% by weight hexagonal crystalline structure and at most 20% by weight rhombohedral crystalline structure.

28. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 2, wherein the temperature of said heat treatment [for transforming] to transform crystalline structure to hexagonal structure is in a range of 900°C to 1100°.

29. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 3, wherein the temperature of said heat treatment [for eliminating] to eliminate impurities is in a range of 2700°C to 2900°C.

30. (Amended) A method [for] of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, comprising the steps of:

providing graphite powder having a particle size equal to or smaller than 100  $\mu\text{m}$ , and having a crystalline structure which is at least 30% by weight rhombohedral crystalline structure and at most 70% by weight hexagonal crystalline structure; and

heating said graphite powder as a heat treatment, or immersing said graphite powder into an acidic solution as an

immersing treatment, to form treated graphite powder, such that the treated graphite powder has a fraction of a hexagonal structure of at least 80% by weight and a fraction of a rhombohedral structure of at most 20% by weight.

31. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 30, wherein the graphite powder has a fraction of a hexagonal structure of at least 90% by weight.

32. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 31, wherein the graphite powder has a fraction of a rhombohedral structure of at most 10% by weight.

34. (Amended) A method [for] of manufacturing graphite powder as claimed in claim 30, wherein the graphite powder is provided by pulverizing raw graphite; and wherein the graphite powder is heated to form treated graphite powder, and after said heating the graphite powder is further heat-treated, at a higher temperature than the temperature of said heating, [for eliminating] to eliminate impurities.

38. (Amended) A method of manufacturing graphite powder adapted to be an active material of a lithium battery negative electrode, comprising the steps of:

providing graphite powder having a particle size equal to or smaller than 100  $\mu\text{m}$ , and having a crystalline structure which is at least 30% by weight rhombohedral

crystalline structure and at most 70% by weight hexagonal  
· crystalline structure; and

treating the graphite powder such that the treated  
graphite powder has a fraction of a hexagonal structure of at  
least 80% by weight and a rhombohedral structure of at most  
20% by weight.